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TITLE: THE INORGANIC INSULATOR PROGRAM AT LASL*

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THE INORGANIC INSULATOR PROGRAM AT LASL*

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## INTRODUCTION

It is the responsibility of this program to address critical and generic materials problems associated with the use of ceramics in fusion reactors. These materials are primarily used as electrical insulators, and the most severe problems result from the presence of radiation fields characteristic of fusion devices. Thus the focus of this work is for the most part on radiation effects in ceramics.

The program is made up of two projects. In the first, experiments are conducted to determine electrical and structural changes resulting from neutron and ionizing radiation. The second involves calculation of damage effects in compounds and the dependence of this damage on neutron energy. These activities are outlined in Sections I and II of this report.

## I. EXPERIMENTAL PROGRAM

Six major applications for ceramics in fusion devices have been identified:(1)

- Insulators for magnetic coils
- The toroidal current break
- Neutral beam injector insulators
- Dielectrics for RF heating systems
- Direct converter insulators
- First wall (including structure, armor, limiters, and collectors).

Those parts of the LASL program that address the use of inorganic insulators for magnets are the subject of this paper.

*Work performed under the auspices of the U. S. Department of Energy.

Six critical and generic ceramic problem areas, three structural and three electrical, are apparent for fusion applications:

- Swelling
- Strength degradation
- Reduction of thermal conductivity
- Decrease in electrical resistivity
- Reduction of dielectric strength
- Increase in loss factor (for RF applications).

All but the last are of concern for magnetic coil applications, and are discussed below.

Swelling

At elevated irradiation temperatures, swelling results from the creation of new lattice sites (e.g., by conversion of displacement-induced interstitials to lattice atoms and stabilization of vacancies in voids). At room temperature and below this mechanism operates less efficiently, but significant swelling can result from lattice dilation induced by finely-dispersed defects. Results of two low-temperature swelling studies conducted at LASL are described here.

$MgO$ and $MgAl_2O_4$ are candidate ceramics for high-dose applications because of their cubic lattice structure (which minimizes problems associated with anisotropic dimensional changes). Samples of these materials were irradiated to fluences of $\sim 2.1 \times 10^{26}$ fast $n/m^2$ ($E > 0.2$ MeV) and $\sim 4.6 \times 10^{26}$ thermal $n/m^2$ at 155°C, generating damage levels which might be encountered in the poorly-shielded divertor coil of a power reactor. Resulting swelling values are:[2]

| | |
|---|---|
| $MgO$ | 2.8 vol % |
| $MgAl_2O_4$ | 0.8 vol %. |

TEM examination showed the defect structure of each to be characterized by fine defect aggregates and dislocation loops. The magnitude of the observed swelling may be sufficient to cause significant dimensional changes in magnetic coils.

MACOR* machinable glass-ceramic is an attractive material for magnet applications, but is sensitive to radiolysis (structural damage induced by ionizing radiation or by the ionizing component of neutron energy loss). Electron irradiation studies near room temperature conducted in the electron microscope show visible structural damage at $\sim 10^{10}$ Gy. However, little or no damage is seen after irradiation at RT to $\sim 10^{22}$ 14 MeV $n/m^2$, corresponding to $\sim 10^7$ Gy, and no swelling is observed.(3,4) If swelling does not worsen at cryogenic temperatures, MACOR would, by the swelling criterion, be useful for such applications as the toroidal field (TF) coil.

## Strength

Strength of ceramics varies directly with fracture toughness, inversely with critical flaw size, and is degraded by internal stresses. Of greatest concern for a polycrystalline ceramic is anisotropic swelling, which usually results from a non-cubic crystal structure or the presence of dissimilar phases. Such swelling can result in high internal stresses and ultimately in microcracking. On the other hand, fracture toughness of ceramics may actually increase, as cracks are pinned by irradiation-induced defects.(5)

Preliminary tensile strength measurements for MgO and $MgAl_2O_4$ irradiated at 155°C (to the fluences described earlier) show the following strength increases:(2)

| | |
|---|---|
| MgO | ~18% |
| $MgAl_2O_4$ | ~21% |

This good performance is encouraging, but it must be recalled that very high stresses can result from swelling unless dimensional changes can be accommodated by coil design.

Bend strength of MACOR after irradiation to $10^{22}$ 14 MeV $n/m^2$ at RT showed essentially no change,(4) consistent with swelling and electron microscopic observations.

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*A product of Corning Glass Works, Corning, NY.

Thermal Conductivity

Heat is primarily conducted by phonons in an electrically-insulating ceramic. Radiation-induced defects scatter phonons and therefore reduce thermal conductivity, with fine-scale damage being the most deleterious at room temperature and above. (At cryogenic temperatures the greater phonon mean free path results in strong scattering by larger, more widely-spaced defects.) Thus degradation of thermal conductivity increases with increasing dose and decreasing irradiation (or measurement) temperature. However, sensitivity to damage is less at higher fluences (Fig. 1).[6] The lower limit for degradation (unless microcracking occurs) is reached when phonon mean free path is reduced to the interatomic spacing (~3 Å), at which point the thermal conductivity approximates that of a glassy structure.

Large decreases in thermal conductivity are anticipated for insulators in divertor coils, where fluences will be high and temperatures relatively low. At the much smaller radiation doses expected beyond the shield (e.g., at TF coils), room-temperature reductions should be small; no decreases were observed in the irradiated MACOR described earlier.[4] However, degradation might be significant at cryogenic temperatures.

Electrical Resistivity

Post-irradiation measurements in $Al_2O_3$ show an increase in electrical resistivity, apparently due to defect-induced scattering or trapping of electronic charge carriers.[7] A slight decrease in resistivity was observed in MACOR after irradiation to $\sim 10^{22}$ n/m$^2$ at RT (Fig. 2),[8] perhaps resulting from changes in ionic conductivity of the glassy matrix.

Resistivity decreases significantly during irradiation, as absorption of ionizing energy enhances the number of charge carriers.[7] This phenomenon is dependent primarily on rate of energy absorption rather than total ionizing dose. Dependence on dose rate is roughly linear in Cr-doped $Al_2O_3$ near RT, although this relationship does not hold at elevated temperatures. Temperature-dependence of radiation-induced conductivity is complex, possibly involving trapping, detrapping, recombination, and/or scattering of charge carriers. The result in $Al_2O_3$ is a lessened temperature dependence (Fig. 3).[7]

A significant reduction in resistivity can be expected in coils subjected to high rates of irradiation, either from gamma or neutron fluxes. Although measurements have not been made at cryogenic temperatures, the low irradiation rate beyond the shield (on the order of 0.1 Gy/s) suggests that radiation-induced conductivity effects will be minimal at that point.

Dielectric Strength

Dielectric breakdown of ceramics occurs by one of two mechanisms, depending on temperature and length of time that voltage is applied. For low temperatures or short times the avalanche mechanism prevails, in which electron collision ionization and multiplication causes breakdown. Under other conditions, thermal breakdown occurs via Joule heating. Cryogenic magnets are expected to operate in the avalanche regime, whereas near-room-temperature coils may be in either the avalanche or thermal regime depending on material chosen and operating conditions.

There is no obvious reason why fine-scale structural damage should degrade the post-irradiation dielectric breakdown strength of a ceramic, unless microcracking occurs. Indeed, no degradation was observed in single-crystal $Al_2O_3$ irradiated to $\sim 2 \times 10^{26}$ $n/m^2$ at elevated temperatures and tested in the avalanche regime (Fig. 4).[9] Defects present included a high concentration of dislocations and voids up to 100 Å dia.[5]

Dielectric strength of polycrystalline $Al_2O_3$ is slightly reduced by an ionizing flux of 6 Gy/s near room temperature, but above ~175°C no degradation was observed.[10] The latter behavior was attributed to a swamping of the ionization contribution by thermal effects. These results suggest that reduction in dielectric strength may not be a problem for shielded magnets, but that close-in coils (e.g., the divertor coil, where the neutron contribution to ionization flux alone will be 1 to 100 Gy/s) may suffer degradation.

## II. DAMAGE CALCULATIONS

The lack of intense 14 MeV neutron sources has made it necessary to conduct high-dose neutron irradiation studies of ceramics in fission reactors. Calculations have been carried out to determine whether fission neutrons supply an adequate simulation of fusion neutrons. Results show that in low-Z ceramics (e.g., $Al_2O_3$, MgO, $MgAl_2O_4$) the relative amount of damage energy deposited in the cation and anion sublattices is roughly the same for fast fission and for fusion neutrons (Fig.5).[11] This is not the case, however with high-Z ceramics (e.g., TaO, $UO_2$), where relative damage levels in the two sublattices differ for the two neutron energies (Fig. 6).[11]

These calculations also show that the ratio of cation to anion displacements can be strongly dependent on displacement thresholds and atomic masses. This observation has important implications for the nature of damage produced and the consequent damage microstructure.

## SUMMARY

1. It does not appear that irradiation problems will be severe for ceramic insulators in well-shielded magnets (e.g., the toroidal field coils), unless materials particularly sensitive to radiolysis are used.

2. Ceramic insulators for poorly-shielded magnets (e.g., divertor coils) could suffer significant degradation of structural and electrical properties, making it important that proper materials choices be made.

3. More data are needed near room temperature for high-dose applications. Of particular importance are:

- fusion neutron data (to be obtained when the Fusion-Materials Irradiation Test Facility becomes available)
- damage results obtained with the proper dpa/gas atom ratio (this can be supplied by irradiation of isotopically-adjusted ceramics in a mixed-spectrum fission reactor)
- measurement of electrical resistivity and dielectric strength during absorption of ionizing energy at appropriate rates.

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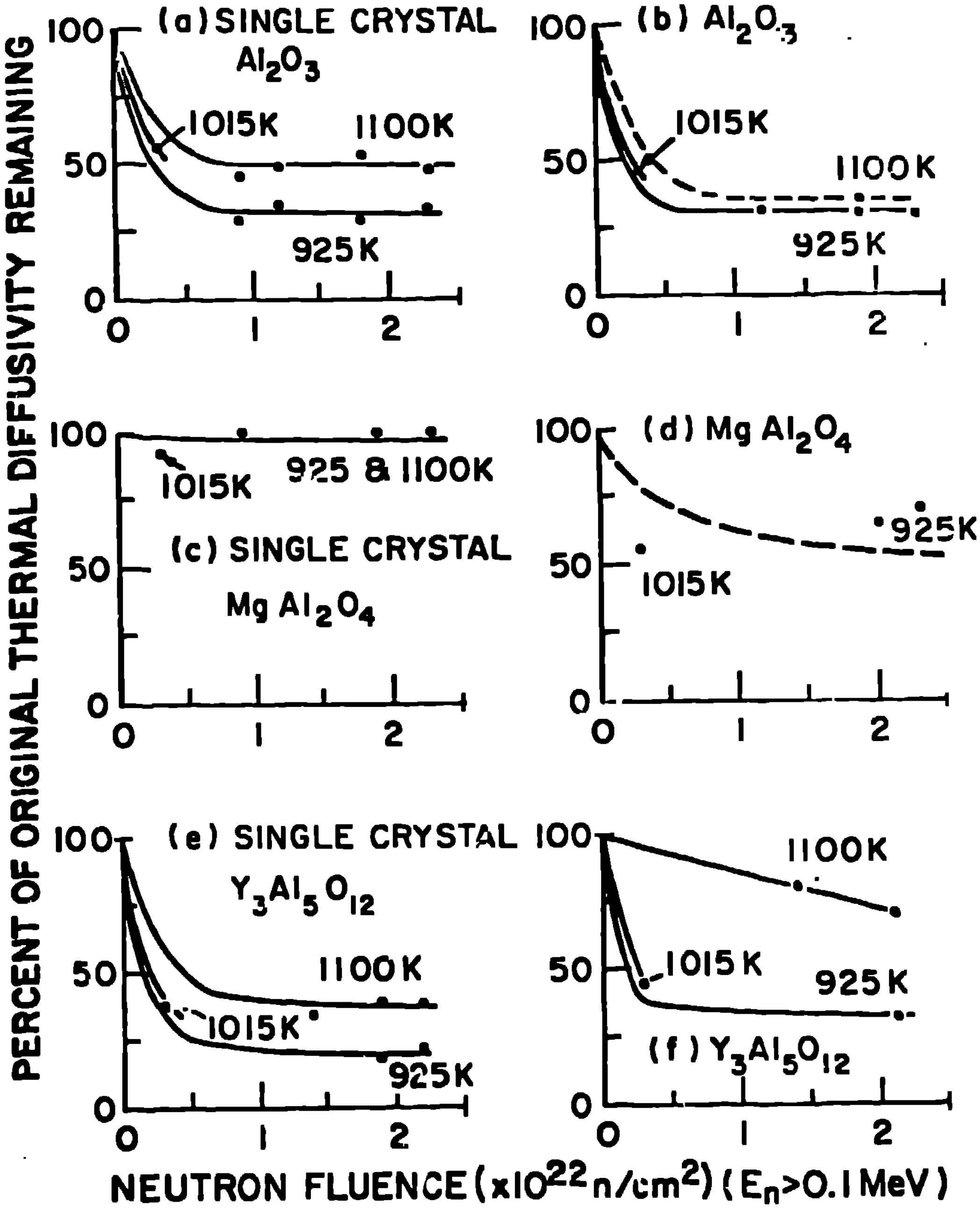


Fig. 1. Decrease in RT thermal diffusivity (approximately proportional to thermal conductivity) as a function of irradiation temperature and fission neutron fluence for several ceramics.[6]

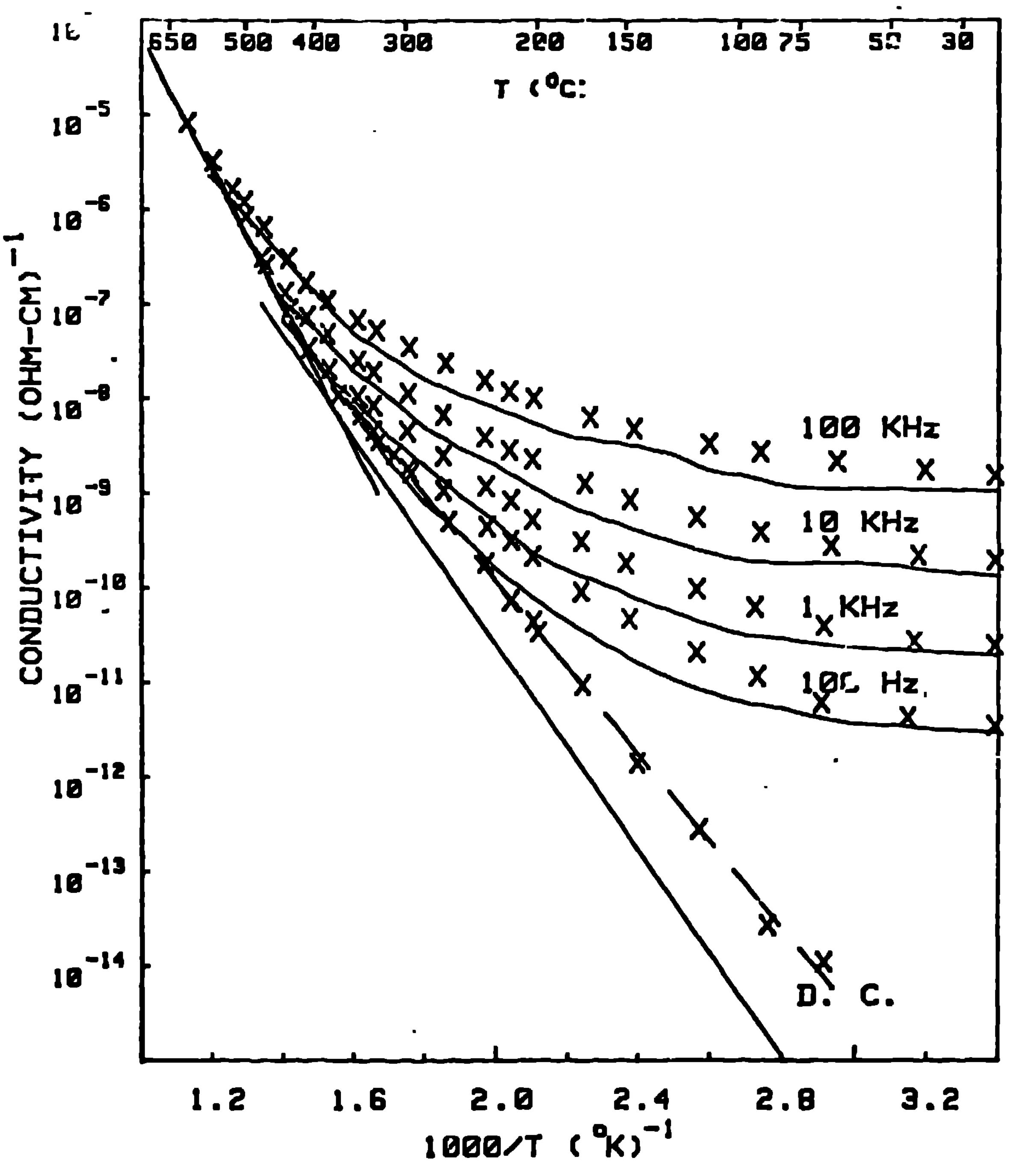


Fig. 2. Electrical conductivity (inverse of resistivity) of MACOR before and after 14 MeV neutron irradiation, as a function of measurement temperature and frequency.(8)

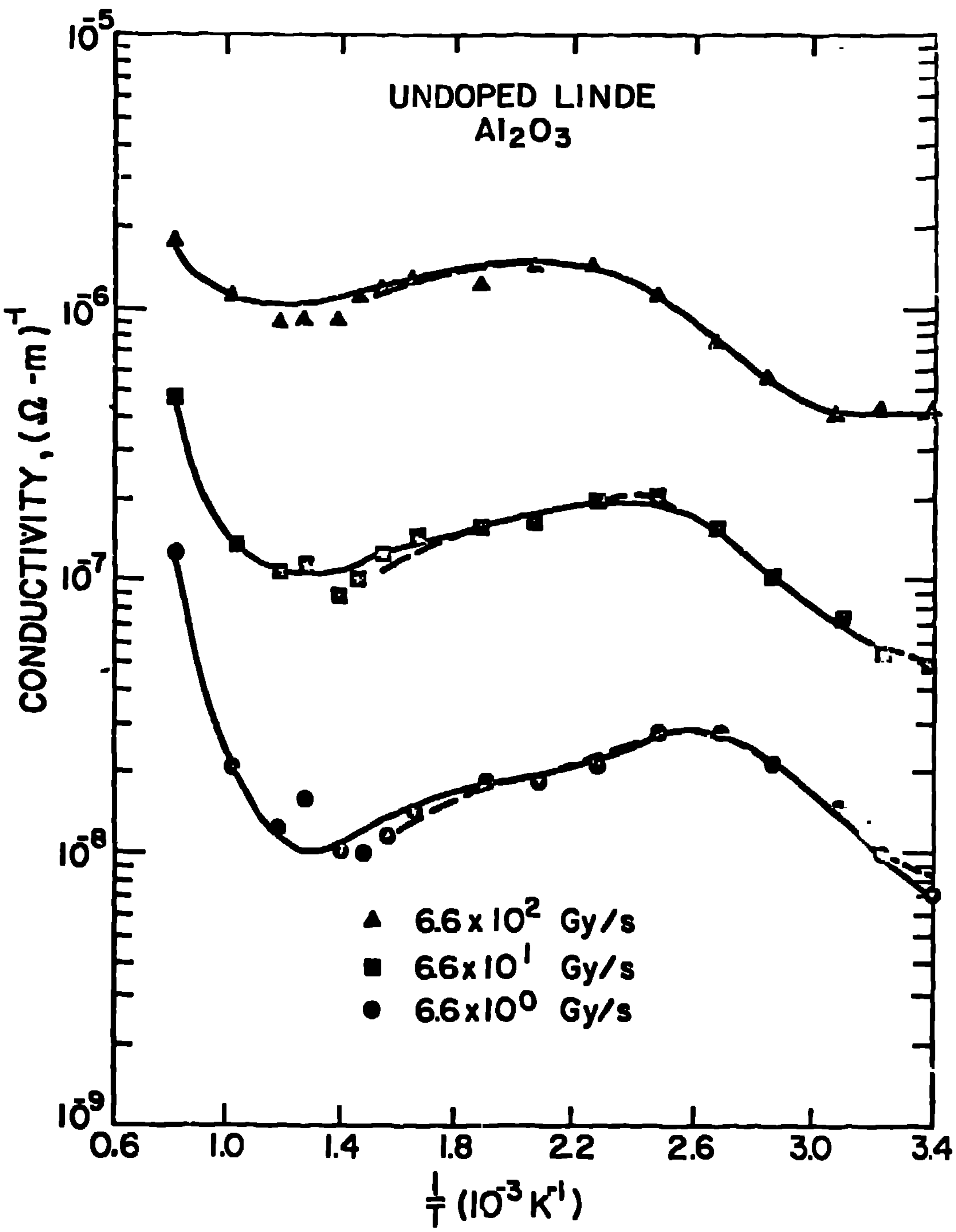


Fig. 3. Dependence of electron irradiation-induced conductivity in single-crystal $Al_2O_3$ on temperature and ionizing dose rate.(7)

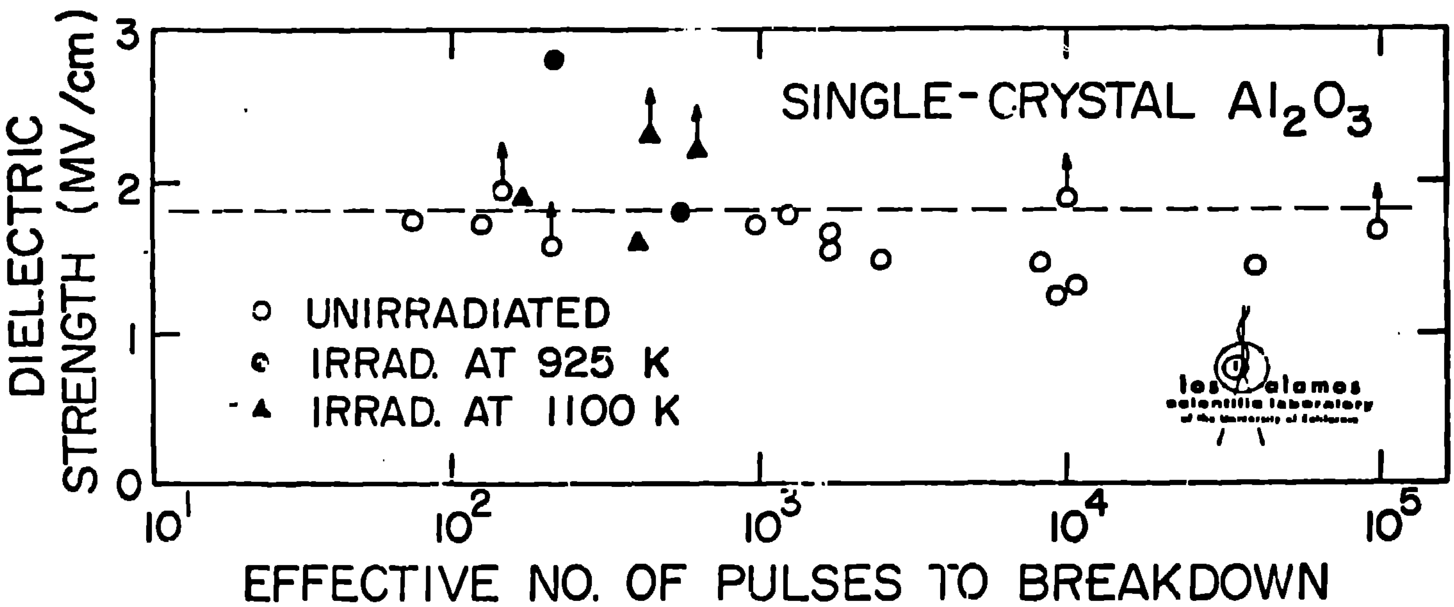


Fig. 4. RT, short-pulse dielectric breakdown strength of $Al_2O_3$ before and after elevated-temperature irradiation to $\sim 2 \times 10^{26}$ n/m$^2$ (E > 0.1 MeV).[9]

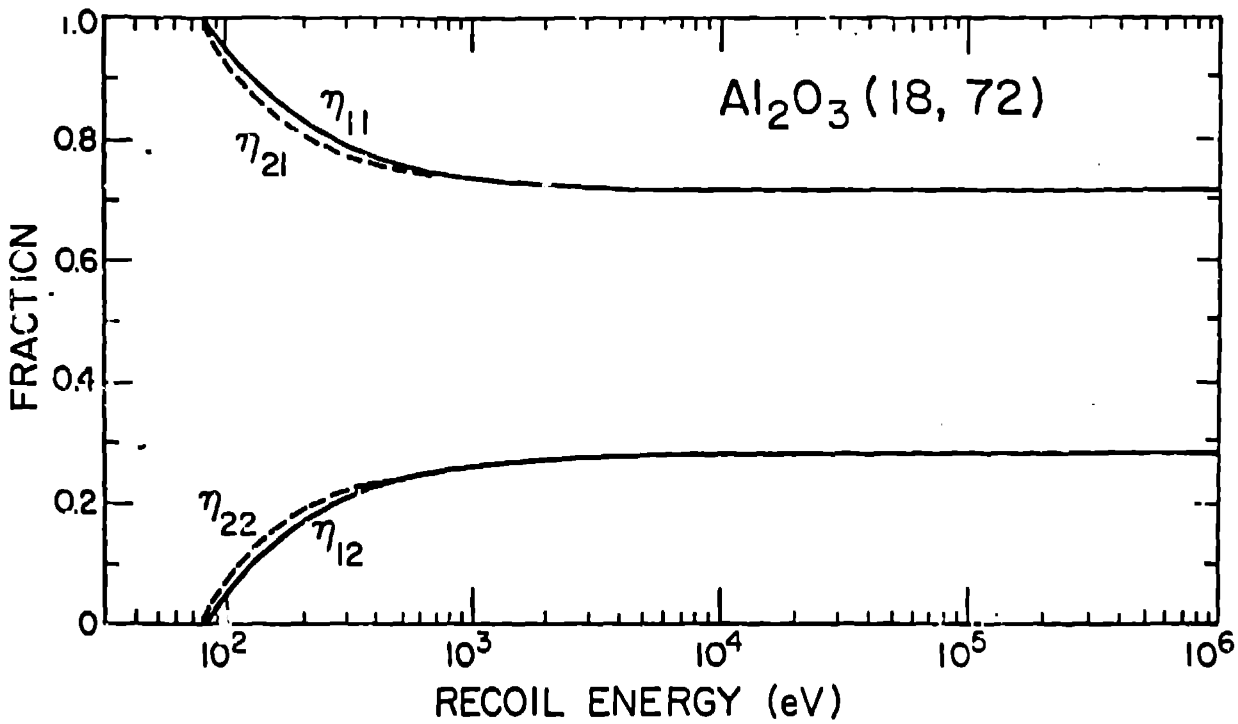


Fig. 5. The fraction $\eta_{ij}$ of type-j displacements produced by a primary knock-on atom of type i in $Al_2O_3$ (Al = 1, O = 2) assuming $E_1{}^d$ = 18 eV, $E_2{}^d$ = 72 eV.[11]

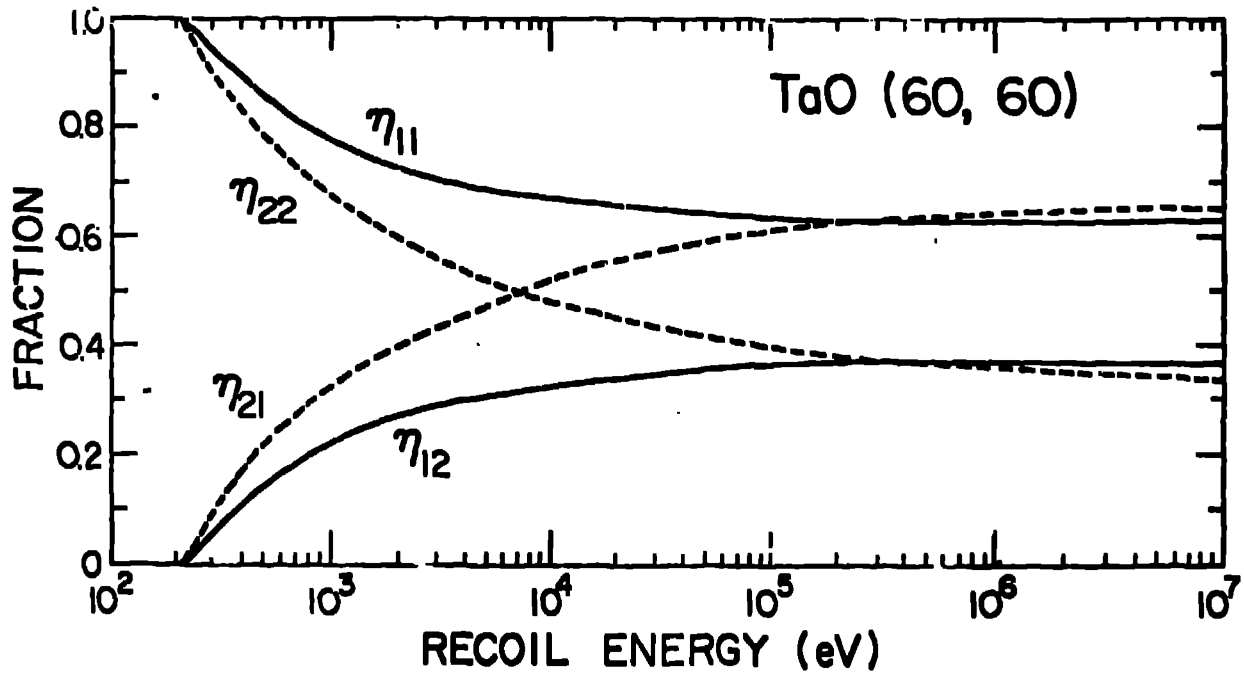


Fig. 6. The fraction $\eta_{ij}$ of type-j displacements produced by a PKA of type i in TaO (Ta = 1, O = 2) assuming $E_1{}^d = E_2{}^d = 60$ eV.(11)